Abstract No. nohe102

**Observation of a monoclinic phase in the piezoelectric system PMN-xPT**

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Beamline(s): X22A

**Introduction**: Different new phases with monoclinic and orthorhombic symmetry have been recently discovered in two technologically important piezoelectric systems, $PbZr_{1-x}Ti_xO_3$ (PZT) and $Pb(Zn_{1/3}Nb_{2/3})_{1-x}Ti_xO_3$ (PZN-x%PT), respectively. These phases are located, in both cases, in between the well-known rhombohedral (R3m) and tetragonal (P4mm) phases, in the temperature vs. composition phase diagrams[1,2]. Such lowering of symmetry is shown to be responsible for the special electromechanical properties observed in these lead complex oxides. In these experiments we have investigated a third lead solid solution known as PMN-x%PT [$Pb(Mb_{1/3}Nb_{2/3})_{1-x}Ti_xO_3$] which is known to behave very similarly to PZN-xPT, that is, showing deformations larger than 1% when the rhombohedral crystals are poled along the <100> directions[3].

**Methods and Materials**: On the one hand, the high lead content of these crystals reduces to a couple of microns the penetration length of typical x-ray energies of about 15keV. On the other hand, the first microns below the surface of the crystal, where the strain induced by the electric field is more easily released, are very different from the rest of the crystal in this type of high piezoelectric materials. High-energy synchrotron x-ray diffraction is thus needed to perform diffraction experiments in transmission mode and to be able to sample the crystal bulk. For this purpose, at X22A we have used 30 keV x-rays by taking advantage of the third order reflection of a Si(111) monochromator. A Si (111) analyzer was used in the diffraction path.

Single crystals of PMN-35%PT were grown from a high temperature solution using the optimum flux composition (49wt%PbO + 1wt%$B_2O_3$)[4]. A platelet 65 μm thick was cut parallel to the reference $(001)_c$ plane and polished with fine diamond paste. The $(001)_c$ faces were covered with Ag-paste, Au-wires were attached and the platelet was poled under an electric field of 43 kV/cm which was applied along [001]cub at 200 $^o$C (above $T_C$ =185 $^o$C) and maintained while cooling down to room temperature.

**Results and conclusions**:

By scanning the reciprocal space around several Bragg reflections in poled PMN-35%PT, we have clearly observed that this material is monoclinic of the $M_A$ type (space group Cm)[5]. Its unit cell is doubled with respect to the tetragonal one and it is rotated 45$^o$ about the c-axis with respect to it, with lattice parameters a= 5.692 Å, b= 5.680 Å, c= 4.050Å and β= 90.15$^o$. The figure shows three contour maps in the reciprocal space around the (002), (300) and (330) pseudo-cubic reflections in the HOL and HHL zones, where the monoclinic domains are evident. The two d-spacings observed around $(330)_c$ correspond to the $a_m$ and $b_m$ monoclinic lattice constants, of 5.692 Å and 5.680 Å, respectively, while the d-spacing observed at $(002)_c$ corresponds to $c_m$= 4.050 Å. The transversal splitting in $(300)_c$ and $(330)_c$ is due to the monoclinic angle β= 90.15$^o$ between $c_m$ and $a_m$. Work is in progress to determine if this $M_A$ phase is also present in the unpoled material (see Abstract No. nohe105).

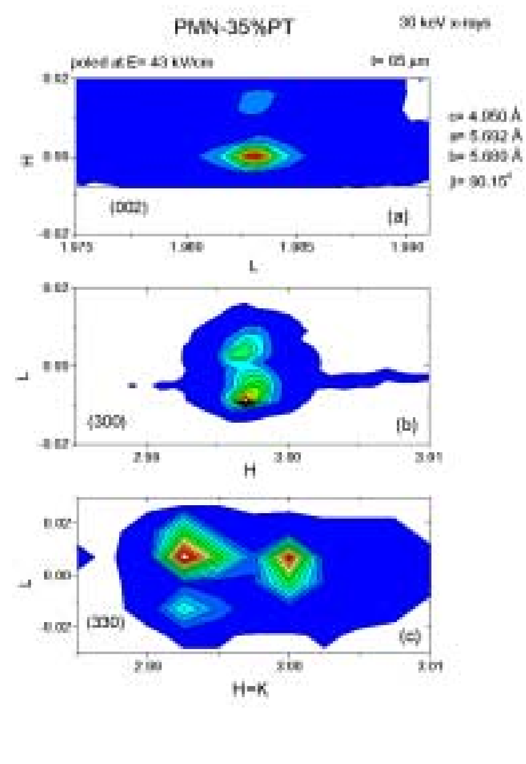


FIG. 1 30keV x-ray diffraction mesh scans around (a) $(002)_c$, (b) $(300)_c$ and (c) $(330)_c$ pseudo-cubic reflections measured in transmission geometry, showing the $M_A$ monoclinic domains (from ref. [5]).

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**References**: [1] B. Noheda et al., Appl. Phys. Lett. **74**, 2059 (1999); [2] D.E. Cox et al., Appl. Phys. Lett. **79**, 400 (2001); [3] S.E. Park and T. Shrout J. Appl. Phys. **82**, 1804 (1997), [4] M. Dong and Z.-G. Ye, J. Cryst. Growth **209**, 81 (2000). [5] Z. Ye et al. Phys. Rev. B **64**, 184114 (2001).